Remote Sensing of Tropospheric Ozone and its Precursors

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Global and regional mapping of Tropospheric ozone and its precursors using the Tropospheric 1 imission Spectrometer and its forerunner - the Airborne 1 imission Spectrometer - will be discussed.

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Introduction

Two closely-related imaging infrared Fourier Transform Spectrometers - the Tropospheric Emission Spectrometer (TES; scheduled for launch on the Earth Observing System CI IEM-1 platform in 2001) and the Airborne Emission Spectrometer (AES; completed in 1994 and flown on a variety of NASA aircraft) - are aimed at elucidating the chemistry of the troposphere on global and regional scales with far better coverage (spatial and temporal) than is feasible with *in situ* sensors during intermittent field and aircraft campaigns,

As was emphasized in a report by the National Academy of Sciences' Board on Sustainable Development (August 1995), the key issue intropospheric chemistry is the formation, transport and interactions of tropospheric ozone. That is, it is insufficient (in our present state of knowledge) merely to monitor ozone itself- its entire chemistry must be investigated. In short-hand form, this is usually stated as "tropospheric ozone and its precursors"; however, a NASA-sponsored workshop at the Goddard Institute of Space Studies in November 1995 pointed out that tile "post-cursors" are equally important and must be included in any program of investigation of tropospheric chemistry.

Tropospheric Ozone and its Precursors

Knowledge of the origin and fate of tropospheric ozone (0₃) is important for several reasons: it is the primary source (through photolysis by solar UV radiation) of hydroxyl radicals (011) which, in turn, provide the mechanism by which pollutants such as carbon monoxide (CO) and methane (CH₄) are removed from the lower atmosphere; through its reactions with hydrocarbons, ozone is the source of PAN (peroxyacetylinitrate), a major ingredient of urban photochemical smog and a powerful lachrymator; ozone itself is hazardous to the health of both plants and animals since it is a powerful oxidant; and, in addition, ozone is a significant greenhouse gas, especially in the upper troposphere.

Thus while *some* tropospheric ozone is essential for human health, too much is deleterious (the EPA allowed maximum is 105 ppbv for any 1 hour per day).

The sources of ozone are two-fold: some is transported down from the Stratosphere (the exact amount being controversial) and some is created in situ through the photolysis of nitrogen dioxide (NO_2) to nitric oxide (NO) which, in turn, reacts with CO and hydrocarbons to form 0. The sum of NO_2 is not consumed in this process (i.e., it is catalytic) and becomes available for further ozone production. Thus urban areas, where combustion and automobile exhausts provide prolific sources of NO_x , are particularly prone to atmospheric pollution (especially in areas of abundant sunlight). However, many ruralareas (for example, the southeastern US) occasionally fail the EPA standard for reasons that are somewhat mysterious. Finally, the sink of NO_x is nitric acid (HNO₃) which is rained out to the surface (and is, incidentally, a component of acid rain).

in order, therefore, to understand tropospheric ozone, one must do more than just measure O_3 itself - it is essential to make co-located measurements of NO_x , $IINO_3$, CO and hydrocarbons (of which CII_4 is by far the most abundant) on a global scale because atmospheric pollution is no respecter of political boundaries. This is the policy issue that the TES and AIS programs have been designed to address.

Measurement Requirements

The species to be measured have their spectral features wide] y scattered throughout the infrared and it therefore follows that the spectrometer must have a wide frequency coverage. The range chosen for both instruments is 650-3050 cm⁻¹. I lowever, in order both to optimize the detector technologies and to control the thermal background radiation, we have chosen to sub-divide the range into 4 regions and further to sub-divide these regions using interchangeable filters 250 - 300 cm⁻¹ wide. In both instruments, 4 bands are observed simultaneously. To improve collection efficiency, each detector package has either 4 (in AES) or 16 (TES) contiguous elements. Thus in any one scan, AES generates 16 simultaneous interferograms and '1'1ES will generate 64.

Only about 10% of the total ozone in the atmosphere resides in the troposphere. The stratospheric component must therefore be discriminated against. 1 fortunately, we are aided by pressure broadening in the lower atmosphere - mar the surface weak lines have a full-width-at-half-maximum of about O. 1 cm⁻¹ whereas stratospheric features are some 10 times narrower. Both TES and Al S have an unapodized spectral resolution of 0.07 cm⁻¹ (TES also has a limb-viewing mode. at ().017 cm⁻¹ resolution; see below).

Unfortunately, some of the critical precursor species (notably the NO_y) have very low abundances (typically parts-per-trillion) and the only way to measure them is to employ the 20x path-length enhancement available by limb viewing. However, this means that measurements can rarely be made below 4 - 5 km (because of clouds) which, in turn, drives us to the higher spectral resolution mentioned earlier because the lines in the upper troposphere are narrower. Most of the rest can be measured in the nadir, where we have a much greater probability of viewing to the surface. Al S dots not have a limb-viewing, capability because an added requirement is imposed by the fact that the limb radiances in the upper t roposphere are very low, so as much of '1'1 S as possible will operate at 180K (the

detectors themselves, and their associated filters and condenser optics are further cooled to 65 K). Such cooling, to 180K is readily achieved on orbit using radiators but it was deemed impracticable (and far too expensive) similarly to cool AES. As a consequence, AES is limited to emulating the TES nadir modes.

The scan speed for TES is held constant for all modes. A nadir observation occupies 4 seconds but the 4x better limb resolution requires 16 seconds. When making our Global Surveys, 2 calibration scam (at 0.1 cm-1 resolution) are followed by 2 nadir scans and 3 limb scans (at the trailing limb). The entire sequence takes about 80 seconds and is repeated continuously for 4 days (57 orbits), followed by 4 days of more extensive calibration and so-called Special Products generation where targetted observations of phenomena such as regional pollution events and volcanic, eruptions can be made. The planned mission lifetime is 5 years.

Data Analysis

TES (and AES) data processing proceeds in 5 steps: at Level 1 A the raw bits out of spacecraft are reconstituted into floating-point interferograms (engineering and housekeeping dala are stripped out and follow a parallel path).] leaders are added (where? when? how?) at this juncture. At 1 evel 1 B the interferograms are phase-corrected, transformed to spectra and the radiometric calibrations ("slope" and "off'-set") applied. These spectral radiances are a deliverable product (i. e., publicly available). Level 2 is the computationally-intensive task of retrieval of concentration profiles. We have chosen to employ the techniques pioneered by Clive Rodgers and his associates, two different implementations of which are currently being used (for AES). The deliverable Level 2 algorithm for TES will incorporate the best features of each. Level 3 (Global Resampling & Mapping) and Level 4 (Global Modelling) are still in definition but will undoubtedly benefit from the on-going activities in 4-D data assimilation at the Goddard Space Flight Center and elsewhere. We expect that our Level 3 maps will be our most widely-used browse product.

Flight Plans

At this writing, TES is about to transition from "concept" phase to "execution" phase in preparation for a Dec 2001 launch. 1 nthe meantime, we have been pursuing an aggressive campaign of flights with Al S:in1994 we made direct, targetted observations of biomass burning (two western wildfires, results in press in JGR); in1995 we participated in the Southern oxidants Study (SOS) Nasbville/Middle Tennessee Intensive Campaign (analysis nearing completion); and in the late fall of 1996 we will participate in the Pacific Rim 1 experiment both as an adjunct to the Thermal Infrared Mapping, Spectrometer volcano observations and to generate extensive transects of tropospheric chemistry over the Pacific Ocean. In the future, we expect Al S to become part of the EOS validation system.